

ATTACHMENT B

Docket No. FE6085 (US)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Diego Brita et al.

Examiner: L.S. CHOI

Serial No. 10/537,077

Group Art Unit: 1796

Filed: June 01, 2005

For: **CATALYST COMPONENTS FOR THE POLYMERIZATION OF OLEFINS**

Hon. Commissioner of Patent and Trademarks
P.O. Box
Alexandria, VA 22313-1450

Sir:

DECLARATION UNDER 37 CFR § 1.132

I, Dr. Tiziano Dall'Occo hereby declare that:

1. I am Dr. Tiziano Dall'Occo.
2. In 1981 I received the Degree of Chemistry from the University of Ferrara in Ferrara, Italy.
3. Since 1984, I have been continuously employed by Basell Poliolefine Italia S.r.l., or its predecessor companies at the Ferrara Research Center in Italy, as a researcher with particular focus in Ziegler-Natta polymerization catalysis. Presently, my specific field of research activity as a senior scientist regards heterogenous Ziegler-Natta catalysts for ethylene polymerization.
4. I have co-authored approximately 36 technical

literature articles, most of them in the field of olefin polymerization. Moreover, I and am the inventor or co-inventor of approximately 35 patents relating to various aspects of catalysts for olefin polymerization.

5. I have studied EP 0 004 647 A2 (herein referred to as "Goeke, et al."), and understand that a generic disclosure of the catalyst, particularly in terms of the Mg/Ti ratio (i.e., from 0.5 to 56) is disclosed. Notwithstanding, the broad Mg/Ti ratio range disclosed, Goeke, et al. expressly teaches the use a much lower ratio of Mg/Ti (i.e., from 1 to 10) in preparing the catalyst precursor, in which the resultant catalyst precursor has a Mg/Ti ratio ranging from 1.5 to 5 (page 16 lines 29-30 in Goeke, et al.). Conversely, in the above-captioned patent application (herein referred to as "the present application"), the molar ratio of the solid catalyst component is higher than 5, and preferably higher than 7, and comprise an unexpectedly higher activity.

6. In order to demonstrate the solid catalyst components comprising a molar ratio of Mg/Ti higher than 5 of the present application comprise unexpectedly higher activities, the attached example and comparative example have been carried out under my supervision. The preparation of the impregnated precursor having formula $\text{TiMg}_3\text{Cl}_{10}(\text{THF})_{6,7}$ (i.e., Example 1a in Goeke, et al.) has been repeated, and is listed as Comparison Example 1 in ATTACHMENT A. The catalyst obtained from the

precursor of Goeke, et al. was preactivated according to the procedure in ATTACHMENT A, and then tested in ethylene copolymerization *vis-à-vis* a catalyst prepared according to the present application, which is listed as an Example of the Invention in ATTACHMENT A. Both the Example of the Invention and the Comparison Example 1 have substantially the same ED/Ti ratio, however, the Example of the Invention comprises a higher Mg/Ti ratio. Additionally, the catalyst prepared according to the present application (i.e., the Example of the Invention) was also preactivated according to the procedure in ATTACHMENT A.

7. Table 1 of ATTACHMENT A clearly demonstrates that the exemplary catalyst prepared according to the present application (i.e., the Example of the Invention) unexpectedly has an activity (Kg/g.ct/h) more than 15 times higher than the catalyst prepared according to Goeke, et al.

8. I further declare that all statements made of my own knowledge are true and that all statements made on information and belief are believed to be true, having been informed that willful false statements and the like are punishable by fine or imprisonment, or both under 18 U.S.C. § 1001, and may jeopardize the validity of the application or any patent issuing thereon.

18/04/2008
Date

Tiziano Dall'Occo
Dr. Tiziano Dall'Occo



ATTACHMENT A

EXAMPLE OF THE INVENTION

Preparation of the solid component

Into a 1 L four-necked round flask, purged with nitrogen, 710 mL of TiCl_4 were introduced at 0°C . Then, at the same temperature, 50 g of a spherical $\text{MgCl}_2/\text{EtOH}$ adduct containing 25 %wt of ethanol (prepared as described in Example 3 of the specification) were added under stirring. The temperature was raised to 130°C in 1 h and maintained for 130 min. Then, the stirring was discontinued, the solid product was allowed to settle and the supernatant liquid was siphoned off. A second TiCl_4 treatment at 110°C (60 minutes) was inserted prior to the washing steps.

The solid was washed six times with anhydrous hexane (5 x 100 mL) at 60°C and once at 25°C . Finally, the solid was dried under vacuum and analyzed ($\text{Ti} = 4.9\%$ wt; $\text{Mg} = 19.5\%$ wt).

In a 500 mL four-necked round flask equipped with a mechanical stirrer and purged with nitrogen, 300 mL of anhydrous hexane and 30 g of the titanium containing solid obtained as disclosed above were charged at room temperature. At the same temperature, under stirring an amount of THF achieve a molar THF/Ti ratio of 8 was added dropwise. The temperature was raised to 50°C and the mixture was stirred for 2 hours. Then, the stirring was discontinued, the solid product was allowed to settle and the supernatant liquid was siphoned off.

The solid was washed 4 times with anhydrous hexane (2 x 100 mL) at 50°C and (2 x 100 mL) at 25°C , recovered, dried under vacuum and analyzed obtaining the following results

- Total titanium	3.2 %	(by weight)
- Mg	12.6%	(by weight)
- THF	32.5 %	(by weight)

Therefore, the Mg/Ti molar ratio in this catalyst component is 7.8 and the THF/Ti molar ratio is 7.4.

In a 500 mL four-necked round flask equipped with a mechanical stirrer and purged with nitrogen, 100 mL of anhydrous hexane and 20 g of the titanium/THF containing solid obtained as disclosed above were charged at room temperature. At the same temperature, under stirring, 31 ml of 20 % wt/vol hexane solution of THA (TriHexylAluminum; $\text{THA}/\text{THF} = 0.25$ m.r.) were slowly added. The stirring was continued for further 30 min and then at the same temperature, 43 ml of 10 % wt/vol hexane solution of DEAC (DiEthylAluminumChloride; $\text{DEAC}/\text{THF} = 0.40$ m.r.) were slowly added. The stirring was continued for 30 min at room temperature and then one hour at 50°C . Finally, the stirring

was discontinued, the solid product was allowed to settle, the supernatant liquid was siphoned off, and the solid was washed three times with anhydrous hexane (3 x 100 mL) at 25°C and dried under vacuum.

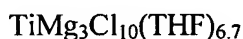
COMPARISON EXAMPLE 1 FROM GOEKE

PREPARATION OF IMPREGNATED PRECURSOR

The example was repeated using a smaller scale but keeping the same molar ratios among the reactants. 26.6 g of Silica (Spherical and porous Grace Silica 952 dried at 800°C for 3 h) were treated, in hexane (140 ml), with 10.8 ml of TEA solution (9.5 % wt/vol), stirred 15 minutes at room temperature and then dried at 60°C under vacuum.

In a 500 mL four-necked round flask equipped with a mechanical stirrer and purged with nitrogen, 128 ml of THF and 2.14 g of anhydrous MgCl_2 were added. Slowly, 0.82 ml of TiCl_4 was dropped and then the mixture was heated to 60°C for 2 hours achieving a complete solution.

The composition isolated by crystallization and analyzed showed a composition in accordance with the formula



At room temperature, the silica/TEA previously obtained, was slowly added and then heated to 60°C and stirred for 2 hours. The suspension was then dried under vacuum at 60°C.

PREACTIVATION:

In a 250 mL four-necked round flask equipped with a mechanical stirrer and purged with nitrogen, 100 mL of anhydrous hexane and 19.1 g of the Silica/titanium/THF containing solid obtained as disclosed above were charged at room temperature. At the same temperature, under stirring, 9.8 ml of 20 % wt/vol hexane solution of THA were slowly added. The stirring was continued for further 30 min and then at the same temperature, 13.2 ml of 10 % wt/vol hexane solution of DEAC were slowly added. The stirring was continued for 30 min at room temperature and then dried under vacuum at 50°C.

Ethylene/1-Butene copolymerization:

A 4.5 liter stainless-steel autoclave equipped with a magnetic stirrer, temperature, pressure indicator, feeding line for ethylene, propane, 1-butene, hydrogen, and a steel vial for the injection of the catalyst, was purified by fluxing pure nitrogen at 70°C for 60 minutes. It was then washed with propane and loaded with 1.14 ml of 10 % wt/vol hexane solution of TEA and heated to 75°C. Finally, 800 g of propane, 1-butene (90 g), ethylene (7.0 bar, partial pressure) and hydrogen (1.2 bar) were charged.

In a 100 cm³ three neck glass flask were introduced in the following order, 5 cm³ of anhydrous hexane, and the amount of solid catalyst reported in table 1. They were mixed together and then introduced in the reactor through the steel vial by using a nitrogen overpressure.

Under continuous stirring, the total pressure was maintained constant at 75°C for 60 minutes by feeding ethylene. At the end the reactor was depressurized and the temperature was dropped to 30°C. The recovered polymer was dried at 70°C under a nitrogen flow and weighted.

Table 1

	Cat. Feed g	Polymer produced g	Specific activity KgPE/g ctz/h	Butene-1 bonded
Comp. Ex. 1	69.8	93	0.7	3.7
Ex. Of the invention	29.5	195	15.9	8.1